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(21) International Application Number: PCT/EP99/07038 (22) International Filing Date: 22 September 1999 (22.09.99) (30) Priority Data: TO98A000800 22 September 1998 (22.09.98) IT (71) Applicant (for all designated States except US): NOVAMONT S.P.A. [IT/IT]; Via Fauser, 8, I-28100 Novara (IT). (72) Inventors; and (75) Inventors/Applicants (for US only): BASTIOLI, Catia [IT/IT]; Via della Noce, 63, I-28100 Novara (IT). BELLOTTI, Vittorio [IT/IT]; Via Mora e Gibin, 9, I-28010 Fontaneto d'Agogna (IT). MONTINO, Alessandro [IT/IT]; Via Bellotti, 15, I-27038 Robbio Lomellina (IT). (74) Agents: RAMBELLI, Paolo et al.; Jacobacci & Perani S.p.A., Corso Regio Parco, 27, I-10152 Torino (IT).	(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>	
(54) Title: POLYMERS OF A HYDROPHOBIC NATURE, FILLED WITH STARCH COMPLEXES (57) Abstract Hydrophobic polymers incompatible with starch containing, as a filler, a starch complex dispersed in the polymer matrix in the form of particles with numeric mean dimension of less than 3 microns, bound to the polymer matrix by coupling agents containing groups compatible with the matrix and with the complex, in which the starch complex is characterized by second-derivative IR absorption in the region of 940-952 cm ⁻¹ are described or wherein the starch complex is bound to the polymeric matrix through reactive groups contained in the complex capable of being fixed to the polymeric matrix. The starch complex in the case of biodegradable polymers such as the aliphatic or aliphatic aromatic polyesters is formed with complexing agents different from the polymer forming the matrix and from EVOH copolymers.		



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INTERNATIONAL SEARCH REPORT

National Application No

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A. CLASSIFICATION OF SUBJECT MATTER IPC 7 C08L29/04 C08L21/00 C08L23/02 C08L67/00 C08L59/00 C08L71/12 C08L69/00 C08K5/548		
According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) IPC 7 C08L C08K		
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Electronic data base consulted during the international search (name of data base and, where practical, search terms used)		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category ²	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 92 14782 A (NOVAMONT SPA) 3 September 1992 (1992-09-03) cited in the application page 19, paragraph 6 -page 20, paragraph 2; claims 1,13 ---	1,6,8,9,20
E	EP 0 965 615 A (NOVAMONT SPA) 22 December 1999 (1999-12-22) claims 1,5,11,12; examples ---	1,12,18
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A	EP 0 404 727 A (WARNER LAMBERT CO) 27 December 1990 (1990-12-27) claims 1,4,11-13; examples --- <div style="text-align: right;">-/--</div>	1,4,7,12
<div style="display: flex; justify-content: space-between;"> <input checked="" type="checkbox"/> Further documents are listed in the continuation of box C. <input checked="" type="checkbox"/> Patent family members are listed in annex. </div>		
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Date of the actual completion of the international search <div style="text-align: center;">25 January 2000</div>		Date of mailing of the international search report <div style="text-align: center;">04/02/2000</div>
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo.nl, Fax: (+31-70) 340-3016		Authorized officer <div style="text-align: center;">Engel, S</div>



INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>DATABASE WPI Derwent Publications Ltd., London, GB; AN 94-057775 XP002031921 & CA 2 069 861 A (BESHAY) 29 November 1993 (1993-11-29) abstract</p> <p>---</p>	1,12,13
X	<p>EP 0 795 581 A (GOODYEAR TIRE & RUBBER) 17 September 1997 (1997-09-17) page 3, paragraph 3; claims 1,3; examples</p> <p>-----</p>	1,13-16, 23



INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

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Polymers of a hydrophobic nature, filled with starch complexes

The present invention relates to polymers of a hydrophobic nature incompatible with starch comprising, as a filler, a starch complex in the form of particles of very small dimensions.

It is known from the literature (WO 92/14782, Bastioli et al. J. of Environmental Pol. Degradation - No. 1, Vol. 3, 181-191, 1993) that starch is present in products produced by the extrusion of mixtures of starch with copolymers of ethylene with vinyl alcohol, in the form of a complex forming an interpenetrated structure with the ethylene copolymer. Upon TEM (Transmission Electron Microscope) examination, the structure shows the presence of phases with sub-micronic dimensions mixed with no sharp separating boundaries.

As a result of treatment in water at 100°C with vigorous stirring, the interpenetrated structure breaks up, forming a micro-dispersion of micro-spherical aggregates with particles of diameter of less than 1 micron, or forming a layered structure by which the starch is rendered partially soluble.

The droplet-like structure has been observed with the use of an EVOH copolymer containing 60% of vinyl alcohol, in moles; the layered structure, on the other hand, has been produced with a copolymer containing 80% of vinyl alcohol, in moles.

There is a great need to be able to disperse starch in polymers incompatible with starch, in the form of a complex which has poor solubility in water, with very small dimensions of the dispersed particles, in view of the improved and novel properties which this filler could introduce.

Up to now, starch has been dispersed in hydrophobic polymers such as polyethylene in the non-destructured, crystalline form.

Destructured starch has been used as a filler in rubbers (USP 5,374,671 and 5,545,680). However, the dimensions of the dispersed particles are not small enough because of difficulties in dispersing the starch finely in an incompatible polymer matrix such as rubber. The starch is in fact dispersed in the form of filament-like particles.

In the United States patents cited above, the possibility of using destructured starch in compositions containing thermoplastic polymers is also pointed out. However, the compositions referred to in the patents are not suitable for the formation of micro-dispersions, both because the method of preparing the compositions is not suitable for the formation of the right micro-structures, and owing to the fact that excessively hydrophilic copolymers unsuitable for forming micro-structures are used. In the case of the EVOH copolymer, the vinyl alcohol content is 73% in moles.

In the patents cited above, reference is also made to the possibility of using a grafting agent which, however, is

not identified further, and which can act as a compatibilizing agent between starch and rubber.

It has now unexpectedly been found that it is possible to disperse, in hydrophobic polymers incompatible with starch, starch complexes characterized by second-derivative FTIR absorption in the region of $940-950\text{ cm}^{-1}$ or by XR diffraction peaks in the regions of $11^{\circ}-13^{\circ}$ and $19^{\circ}-21^{\circ}$ of 2θ , in the form of particles with poor solubility in water and having numeral average size of less than 3 microns, preferably less than 1 micron, and which are fixed to the polymer matrix by means of (a) coupling agent(s) containing groups which can interact with the polymer matrix and with the complex (external coupling agent) or by means of reactive groups present in the complex capable of being fixed to the polymeric matrix thus acting as internal coupling agent. In this case, the use of the external coupling agent can be omitted.

This is, for example, the case of matrices comprising a biodegradable polymer such as the aliphatic or aliphatic-aromatic polyesters, the aliphatic polyamides, polyamides-polyesters, polyurethane-polyesters and the like.

As it will be specified hereinafter, the complex usable with the biodegradable matrices of the type above indicated, is a complex of starch with a polymer different from the polymer forming the polymeric matrix and from the ethylene-vinylalcohol copolymers, or with other complexing agents.

The complex is generally substantially insoluble in water at 100°C.

The solubility is generally less than 20% by weight.

Amylose is present in the complex in wholly or largely complexed form, whereas the amylopectin can be hydrolyzed with acids. It is considered, but this is not binding, that the amylopectin macro-molecules are connected at various points of the chain by hydrogen bonds and by entanglements with the molecules of amylose complexed with the synthetic polymer or other complexing agents. Since the dimensions of the micro-particles of the complex are of the order of those expected for the amylopectin molecules, these molecules can be considered as forming a nucleus surrounded by a shell formed by the amylose/synthetic polymer complex or other complexing agent.

The shell, by interacting with hydrogen bridges or by entanglements with the nucleus, acts as a screen to the solvation of the amylopectin.

The starch compositions used for the dispersion indicated above are constituted by or comprise starch complexes showing the above specified FTIR band or XR diffraction, peaks from which micro-dispersions of particles with numeral average diameter of less than 1 micron are formed by treatment with boiling in water with vigorous stirring.

The starch usable for the formation of the complexes usable as fillers contains more than 15% by weight, and preferably more than 20% by weight, of amylose; amylopectin is

present in quantities up to 85% by weight and preferably up to 80%.

The starch may originate from tubers, cereals or beans and may be maize, potato, tapioca, pea, or rice starch, etc. It is preferably starch with an amylose content greater than 20% by weight.

Starches with an amylopectin content greater than 85% by weight are not suitable since the amylopectin does not form complexes with the polymers which complex with amylose; a large quantity of the starch is solubilized by treatment by boiling in water.

The compositions mentioned above are prepared by extrusion of the starch in mixture with the complexing thermoplastic polymer and, optionally, with a plasticizer under temperature and shear-force conditions such as to render the components of the mixture rheologically compatible or with a complexing agent. Suitable preparation methods are described, for example, in WO 92/14782, which is incorporated by way of reference. It is also possible to use methods in solution, using common solvents for starch and the complexing agent.

The compositions preferably have a water content at the output of the extruder, before conditioning, of less than 20% by weight and preferably less than 10% by weight, but preferably not lower than 2% and preferably not lower than 4%. A preferred water content as referred to starch plus water in the initial formulation is higher than 5% and

lower than 30%. The Tg of the compositions is preferably below 0°C.

The formation of micro-dispersions by boiling in water with stirring and optionally ultrasonication can constitute a criterion for the selection of the operating conditions suitable for the formation of the compositions usable in the method of the invention.

The polymer compatible with starch contains hydrophilic groups intercalated with hydrophobic sequences in which the hydrophilicity properties are balanced in a manner such that the resulting extruded compositions can provide a partial or complete insolubilization of the starch by treatment in boiling water.

For example, in the case of copolymers of ethylene with vinyl alcohol, the vinyl alcohol content, which is preferably greater than 50% in moles, has not to exceed 80-90% in moles, otherwise, by boiling in water, the formation of layers instead of micro-dispersions occurs and starch becomes soluble.

Other suitable copolymers are copolymers of ethylene with acrylic acid, preferably containing from 15 to 25% by weight of acrylic acid.

In general, all copolymers of ethylene with polar monomers such as copolymers with methacrylic, crotonic and itaconic, acids, maleic anhydride and terpolymers containing vinyl acetate are suitable.

Other polymers which can form complexes with starch are 6-6, 6-9 or 12 aliphatic polyamides, aliphatic and aliphatic-aromatic polyesters, polyurethanes/polyamides, polyurethanes/polyethers, polyamides/polyesters, polyurea/polyesters, polyurea/polyethers, polylactic acid, polyglycolic acid, poly(lactic-glycolic) acid, polycaprolactone/urethane, in which the size of the polycaprolactone block is between 300 and 3000 molecular weight.

Other complexing agents can be fatty acids and their derivatives. The complexing agents can contain reactive groups for the hydrophobic matrix thus performing the function of internal coupling agents, such as tetrasulphide or unsaturated groups in case of rubbery matrices.

In the case of ethylene/vinyl alcohol copolymers containing from 20 to 50% of ethylene, in moles, the quantity of copolymer which can complex all of the available amylose is about 20% by weight of the composition.

The more the concentration of the EVOH is reduced, the greater is the relative quantity of amylose complexed, this quantity changing from twice the quantity of EVOH when the concentration of EVOH is 10%, to 3 times when it is reduced to 5%.

This shows that complexing between amylose and EVOH does not form a complex of well-defined composition, but forms a family of complexes.

The preferred formulations for the starch complexes comprise a content by weight of starch between 45 and 65%, with more than 20% of amylose; a complexing agent between 5 and 35%; plasticizers from 0 to 20% and added water from 0 to 15%. Such formulations minimize the size of the dispersed phase.

The dispersion of the composition comprising the starch complex in the hydrophobic polymer is performed by blending in accordance with known methods, for example, by extrusion or calendering in a Banbury mixer in the case of rubbers.

The preferred complexed starch for rubbery compositions is dispersable in the rubber by mixing in a range of temperature between 130 and 170 °C, preferably between 140 and 160 °C.

It is possible to operate in the presence of a coupling agent. When the complex contains groups which can be fixed to the polymer matrix of the starch, the use of the coupling agent can be omitted.

Suitable coupling agents which can react with the filler and with the polymer matrix when the matrix is of a polyolefin nature or is a styrene-butadiene, polybutadiene, polyisoprene or nitrile rubber, an elastomeric, ethylene/propylene or ethylene/propylene diene copolymer are aliphatic silanes such as dimethyldichlorosilane, methyltrichlorosilane, mercaptopropyltrimethoxysilane and vinyl silanes such as methacryl-oxy-propyltrimethoxysilane and vinyltriethoxysilane.

A particularly suitable silane is bis-3-triethoxysilylpropyl tetrasulphide.

Other coupling agents which may be used are alkyl titanates or esters such as tetraisooctyl titanate, isopropyl-diisostearyl-metacryl titanate, and isopropyltriacyl titanate.

The quantity of coupling agent is between 0.05 and 10% by weight of the starch complex, preferably 0.1-5% by weight.

The coupling agent is preferably added to the starch complex/hydrophobic polymer mixture at the stage of the blending thereof.

The hydrophobic polymer comprises, among others, ethylene polymers such as LDPE, LLDPE, HDPE, ultra low LLDPE, crystalline propylene polymers and copolymers, in particular, isotactic polypropylene, and crystalline propylene copolymers containing 1-10% by weight of ethylene or of a C₄-C₁₀ alpha olefin.

Other thermoplastic hydrophobic polymers which may be used comprise polyamides, aromatic polyester resins, polyoxymethylene resins, polycarbonates, polyphenylene oxide resins. The rubbers used in the tire industry, such as styrene-butadiene rubbers, polybutadiene or polyisoprene rubbers, or the EP and EPDM rubbers may also be used.

The rubbers may contain, as fillers, the ingredients such as, for example, silica, carbon black and the vulcanizing agents and vulcanization accelerators which are normally

used in this field. The rubber matrices containing the starch complexes according to the present invention are conveniently used in the preparation of tyres having valuable properties.

Similarly, the thermoplastic polymers may contain additives such as colourings, stabilizers, and flame-retardant compounds normally used in the field.

The fillers having the characteristics specified above confer to the polymer matrices properties of better coatability, particularly in the case of olefin polymers, better electrical and heat dissipation, a better elastic flow and low hysteresis (particularly in the case of rubbers), and other advantageous properties which vary from one polymer to another.

Biodegradable hydrophobic polymers are also usable. Examples of such polymers are: aliphatic, polyesters, aliphatic-aromatic copolyesters, aliphatic polyamides, polyamides-polyesters polyurea-polyesters, polyurethane-polyesters poliurethane-polyamide. Specific examples are poly-epsilon-caprolactone and poly(butylene terephthalate-butylene adipate).

In the case of the above mentioned biodegradable polymers it has been found advantageous to add to the polymeric matrix a starch complex wherein the complexing agent is a polymer different from the polymer forming the matrix and the ethylene-vinylalcohol copolymers, or selected from the fatty acids and the derivatives thereof or from other complexing agents.

It is possible using this type of complex to tailor-make the properties of starch to render the same similar to those of the matrix or different.

For example in the case of the compostable bags, it has been found useful to retard the biodegradability of the starch complex dispersed in the aliphatic or aliphatic-aromatic polyester matrix by complexing the starch with a slowly or not at all biodegradable polymer to avoid the premature degradation of the bag when the same, filled with waste, is stored in humid places and/or contact with condense.

The premature bag breakage with consequent waste spreading is troublesome and represents a limit to the use of the compostable bags for the collection of organic waste.

The polymers used to retard the biodegradability of starch comprise polylactic acid, polyglycolic acid and poly(lactic-glycolic) acid copolymers.

Examples of not significantly biodegradable polymers are the ethylene-vinylacetate copolymers, the ethylene-acrylic acid copolymers, and in general, the ethylene copolymers containing polar groups different from the OH groups.

In the case of complexing agents immiscible with the polyester base-matrix, it is possible to form the complex with the starch directly during the melt-blending of the polyester-starch mixture.

The melt-blending is carried out under temperature and shear conditions suitable to render the starch and the polyester polymeric components rheologically compatible.

As already mentioned, the dispersion of the starch complex in a matrix formed or comprising a biodegradable polymer such as the aliphatic or the aliphatic-aromatic polyesters does not require the use of an external coupling agent.

The filler formed by the starch complex is dispersed in the hydrophobic polymer in quantities of from 0.5 to 50% by weight. The most suitable quantity depends on the type of polymer and on the properties to be imparted thereto. In general, quantities of from 2 to 30% by weight may advantageously be used.

For the test with boiling water, the material is ground in a cryogenic mill and reduced to powder which can pass through 0.5 mm mesh.

The powder is introduced into a reflux flask containing a volume of water 10 times the weight of the powder and is heated to boiling point for 4 hours with vigorous stirring and ultrasonication, if needed.

The following examples are provided by way of non-limiting illustration of the invention.

In the following examples, the quantities of the components are expressed as percentages by weight unless specifically indicated otherwise.

EXAMPLE 1

A mixture was prepared, containing:

- 40% Cerestar Globe 03401 starch (12.8% water)
- 40% Nippon Gohsei A-4412 EVOH (EVOH with 44% ethylene by moles)
- 12% glycerol
- 3% water
- 5% urea.

The mixture was supplied to an OMC single-screw extruder with D=20 mm and L/D=30, operating with the following temperature profile: 80/150/140/120°C and about 40 rpm..

The extruded material with about 6.5% H₂O was pelletized and then filmed by blow extrusion with a Haake extruder with D=10 mm and L/D=20 to give a film about 30 microns thick.

The film produced was ground in a cryogenic mill and reduced to powder which could pass through 0.5 mm mesh. About one gram of powder was then poured into a flask containing 100 ml of distilled water and the mixture was brought to boiling point with vigorous stirring for 4 hours. Upon completion, the insoluble residue, which was about 75% of the initial quantity, which amount corresponded to the sum of starch and EVOH, was filtered out.

The boiling residue, examined by TEM (Transmission Electron Microscope), was constituted by individual particles or by

aggregates of particles in which the individual particles had diameters of less than 0.5 microns.

The film, examined by second derivative IR had a band of the starch/EVOH complex at 947 cm^{-1} and two peaks in the XR diffraction spectrum at 13° and 20° of 2θ .

EXAMPLES 2-4

The following compositions, in which maize starch containing about 28% of amylose was used, were prepared as described in Example 1:

Example	2	3	4
	%	%	%
Cerestar Globe 03401 starch	50	58	65
Nippon Gohsei E-3808 EVOH	30	20	10
Glycerol	7	8	9
Urea	4	4	4
Water	9	10	12

(EVOH E-3808 with 38% ethylene by moles)

Upon application of the breakdown upon boiling test, amounts of insoluble residues approximately equal to the sum of starch and EVOH were obtained. The dimension of the separated particles was less than 0.5 microns.

All of the films, when examined by second-derivative IR, had a band of the complex at 947 cm^{-1} and when examined by XR showed two peaks at about 13° and 20° of 2θ .

EXAMPLES 5-16

The following compositions, in which a starch with a high amylose content (Roquette Eurylon 7, 70% amylose) was used, were prepared as described in Example 1:

Example	5	6	7	8	9	10	11	12	13	14	15	16
Eurylon 7	52	60	66	70	52	60	66	70	51	60	66	70
D-2908 EVOH	31	20	10	5	-	-	-	-	-	-	-	-
E-3808 EVOH	-	-	-	-	31	20	10	5	-	-	-	-
A-4412 EVOH	-	-	-	-	-	-	-	-	31	20	10	5
Glycerol	7	10	12	12	7	10	12	12	7	10	12	12
Water	10	10	13	13	10	10	12	13	10	10	12	13

Upon application of the breakdown in boiling water test, amounts of insoluble residues equal to the sum of starch and EVOH were obtained, even with the lowest concentrations of EVOH. The size of the separated particles was less than 1 micron, in all the cases but for the concentration of Eurylon between 52 and 60% the particles were by far lower than 0.5 microns.

All of the films, when examined by second-derivative IR, had a band of the complex at 947 cm^{-1} .

EXAMPLE 17

Products obtained according to examples 2,3 and 4 brought to complexed starch with a water content between 6 and 10% which was mixed in a Pomini Farrel mixer at 155C at a content of 20% by weight, with 76% of an SB standard grade for treads and 4% of bis-3-triethoxysilyl propyl tetrasulphide. The final products after etching in DMSO

showed a microdispersion of complexed starch with average dimension lower than 0.5 microns under TEM analysis.

COMPARISON EXAMPLE 1

A composition similar to that of Example 5 but with the maize starch replaced by amylose-free waxy starch (Snowflake 04201 - Cerestar) was prepared.

Filming of the composition and its subjection to the breakdown in boiling water test did not produce a dispersion of micrometric particles, but a quantity polymer lumps equal to the quantity of EVOH present in the molecule.

A portion of the film examined by TEM showed a micro-layered structure.

COMPARISON EXAMPLE 2

Comparison Example 1 was repeated but with 7.1 parts of waxy starch replaced by Eurylon 7 starch so that the final mixture had an amylose concentration of 5%. Filming of the composition and its subjection to the breakdown in boiling water test produced a residue corresponding to 40 parts of EVOH and 15 parts of starch; this means that the quantity of amylose present was not sufficient to screen the solvation of all of the amylopectin.

A portion of the film, examined by TEM, was shown to be of micro-layered structure.

COMPARISON EXAMPLE 3

Example 1 was repeated but with the EVOH replaced by Du Pont Elvanol 71-30 polyvinyl alcohol.

Filming of the composition and its subjection to the breakdown in boiling water test did not produce any insoluble residue.

A portion of film, examined by TEM, showed a micro-layered structure.

EXAMPLE 18

The following composition (parts by weight):

	<u>Comparison</u>	<u>A</u>	<u>B</u>
Maize starch	26.4	26.4	26.4
Ecoflex (BASF)	63.8	53.3	56.3
Eco-PLA D4200 (Cargill)	0	7.5	0
Lactic/glycolic acid copolymer (70:30)	-	-	7.5
Glycerine	5.5	4.35	4.6
Water	4.3	3.45	3.5

(Ecoflex is a poly(butylene adipate/terephthalate) copolymer) were extruded in a twin-screw extruder APV 2030 operating at 180°C/160 rpm and with a throughput of 40 Kg/h.

The water content and MFR after extrusion were:

	<u>water</u>	<u>MFR (dg/min)</u>
Comparison	1.6%	2.3
Test A	1.7%	2.6
Test B	1.6%	2.5

The material was filmed in a Ghioldi apparatus (40 mm diameter and head of 100 mm)

Bags (60 X 90 cm) were produced.

A test using cut grass to fill the bags, conducted at 30°C and 75% RH gave the following result expressed as days for obtaining non-transportable bags:

	<u>days</u>
comparison	3
Test A	20
Test B	23

The IR spectrum of the film gave a FTIR band at about 947 cm^{-1} typical of complexed starch.

After dissolution of Ecoflex, the residual starch was found to be complexed with polylactic acid and poly(lactic-glycolic) acid.

CLAIMS

1. Hydrophobic polymers incompatible with starch containing, as a filler, a starch complex dispersed in the hydrophobic polymeric matrix in the form of particles with a numeral average size of less than 3 microns, bound to the polymer matrix by means of coupling agents containing groups compatible with the matrix and with the starch complex or by means of reactive groups present in the starch-complex capable of being fixed to the polymeric matrix, wherein the starch complex is characterized by a second-derivative IR absorption in the region of $940-950\text{ cm}^{-1}$, and the starch complex, in the case of hydrophobic biodegradable polymers selected from the group consisting of the aliphatic or aliphatic-aromatic polyesters, the aliphatic polyamides, polyamides-polyesters, polyurethane polyesters, polyurethane-polyamides, polyurea-polyesters is a complex of starch with a polymer different from the polymer forming the matrix and from ethylene-vinylalcohol copolymers or with other complexing agents.

2. Polymers according to Claim 1, in which the starch complex is dispersed in the form of particles having a numeral average size of less than 1 micron.

3. Polymers according to Claims 1 or 2, wherein the coupling agent is selected from the group consisting of a vinyl silane, an alkyl titanate, and bis-3-triethoxysilylpropyl tetrasulphide.

4. Polymers according to Claims 1 or 2, wherein the complexing agent different from the polymer forming the

polymeric matrix is selected from the group consisting of polylactic acid, polyglycolic acid, poly(lactic-glycolic) acid copolymers, ethylene-acrylic acid copolymers, ethylene-vinylacetate copolymers.

5. Polymers according to any of Claims 1 to 4, in which the quantity of filler comprising the complex dispersed in the hydrophobic polymer is from 0.5 to 50% by weight.

6. Polymers according to any of Claims 1 to 5, in which the starch complexes are produced from compositions of starch with polymers compatible with starch containing lyophilic groups and lyophobic sequences, wherein the starch complex is present and from which a micro-dispersion of particles with numeral average diameters of less than 1 micron is formed by treatment in water at 100°C under stirring.

7. Polymers according to any of Claims 1 to 6, produced with the use of compositions having a water content of less than 20%, and higher than 2% by weight, and a Tg below 0°C.

8. Polymers according to Claim 6, in which the polymer which can form complexes with starch is selected from the group comprising copolymers of ethylene with polar monomers.

9. Polymers according to Claim 8, in which the copolymer is selected from the group comprising copolymers of ethylene with vinyl alcohol, vinyl acetate and acrylic acid.

10. Polymers according to Claim 9, in which the ethylene/vinyl alcohol copolymer contains from 50 to 75% of vinyl alcohol in moles.

11. Polymers according to Claim 6, in which the polymer which can complex with the starch is selected from copolymers of polyester/polyurethane, polyamide/polyester, aliphatic and aliphatic aromatic polyesters and polyamides.

12. Polymers according to any of Claims 1 to 11, in which the hydrophobic polymer incompatible with starch is selected from the group consisting of ethylene polymers and copolymers, crystalline propylene polymers and copolymers, aromatic polyester resins, polyamides, polyoxymethylene resins, polyphenylene oxide resins, and polycarbonates.

13. Polymers according to any of Claims 1 to 11, in which the hydrophobic polymer is a rubber selected from the group consisting of styrene-butadiene rubbers, polybutadiene rubbers, polyisoprene rubbers, ethylene-propylene and ethylene-propylene-diene rubbers, and natural rubber.

14. A method for preparing filled polymers according to any of Claims 1 to 13, in which a composition comprising the starch/polymer complex, forming part of a continuous interpenetrated structure between the complexing polymer and the complex is mixed, in the melt state or under hot mastication conditions, with the hydrophobic polymer incompatible with starch, in the presence of coupling agents containing groups reactive with the polymer matrix and with the complex.

15. A method of preparing filled polymers according to any of Claims 1 to 13, in which a composition comprising the starch/polymer complex is mixed with a rubber at a processing temperature between 140 and 160C, in the presence of coupling agents containing groups reactive with the polymer matrix and with the complex.

16. A method according to Claim 14 and 15, in which the coupling agent is selected from vinyl and tetrasulphide silanes and alkyl titanates.

17. A method according to any of claims 14, 15 and 16 in which the coupling agent is used in a quantity of from 0.05 to 10% by weight of the complex.

18. A method for preparing filled polymers according to Claims 1 to 13, wherein the polymeric matrix is a biodegradable polymer selected from the group consisting of the aliphatic-aromatic polyesters, the aliphatic polyamides, the polyamides-polyesters, polyurethane-polyesters, polyurethane-polyamides and poliurea-polyesters comprising melt-mixing the polymer forming the polymeric matrix with a complex of starch having the characteristics as set forth in claim 1 and further characterized by being formed of starch complexed with a polymer different from the polymer forming the matrix and from the ethylene-vinylalcohol copolymers or with other complexing agents.

19. A method according to claim 18, wherein the starch complex is preformed or formed during melt-mixing.

20. Shaped articles obtainable from the hydrophobic polymers of claim 1 to 13.

21. Shaped articles obtainable from the hydrophobic polymers of claims 1 to 13, wherein the hydrophobic polymer is selected from the group consisting of the aliphatic and aliphatic-aromatic polyesters, polyurethane-polyamides, polyurea-polyesters, and polyurethane-polyesters.

22. Films and compostable bags obtainable from the hydrophobic polymers of claims 1 to 13.

23. Tyres obtainable from the rubbers of claim 13.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 99/07038

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08L29/04 C08L21/00 C08L23/02 C08L67/00 C08L59/00
C08L71/12 C08L69/00 C08K5/548

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08L C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 92 14782 A (NOVAMONT SPA) 3 September 1992 (1992-09-03) cited in the application page 19, paragraph 6 -page 20, paragraph 2; claims 1,13	1,6,8,9, 20
E	EP 0 965 615 A (NOVAMONT SPA) 22 December 1999 (1999-12-22) claims 1,5,11,12; examples	1,12,18
X	WO 98 20073 A (BELLOTTI VITTORIO ;CELLA GIAN DOMENICO (IT); DEL GIUDICE LUCIANO () 14 May 1998 (1998-05-14) claims 1,11,28,38,40,48	1,2,4, 12,18,22
A	EP 0 404 727 A (WARNER LAMBERT CO) 27 December 1990 (1990-12-27) claims 1,4,11-13; examples	1,4,7,12
-/-		

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- "&" document member of the same patent family

Date of the actual completion of the international search

25 January 2000

Date of mailing of the international search report

04/02/2000

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Authorized officer

Engel, S

INTERNATIONAL SEARCH REPORT

Int. l. Application No
PCT/EP 99/07038

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages.	Relevant to claim No.
X	<p>DATABASE WPI Derwent Publications Ltd., London, GB; AN 94-057775 XP002031921 & CA 2 069 861 A (BESHAY) 29 November 1993 (1993-11-29) abstract</p>	1, 12, 13
X	<p>EP 0 795 581 A (GOODYEAR TIRE & RUBBER) 17 September 1997 (1997-09-17) page 3, paragraph 3; claims 1,3; examples</p>	1, 13-16, 23

INTERNATIONAL SEARCH REPORT

Information on patent family members

In International Application No

PCT/EP 99/07038

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9214782 A	03-09-1992	IT 1245408 B	20-09-1994
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		YU 118090 A	31-10-1991

REC'D 27 DEC 2000

WIPO

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference PC273PR	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No. PCT/EP99/07038	International filing date (day/month/year) 22/09/1999	Priority date (day/month/year) 22/09/1998
International Patent Classification (IPC) or national classification and IPC C08L29/04		
Applicant NOVAMONT S.P.A. et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 4 sheets, including this cover sheet.

- ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 5 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand 18/04/2000	Date of completion of this report 21. 12. 00
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Boletti, C Telephone No. +49 89 2399 8527 



**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/EP99/07038

I. Basis of the report

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).)*:

Description, pages:

1,2,4-18	as originally filed	
3	with telefax of	19/10/2000

Claims, No.:

1-24	with telefax of	19/10/2000
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2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:



**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/EP99/07038

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims
	No:	Claims 1
Inventive step (IS)	Yes:	Claims
	No:	Claims 2-24
Industrial applicability (IA)	Yes:	Claims 1-24
	No:	Claims

2. Citations and explanations
see separate sheet



**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/EP99/07038

ITEM V

Reference is made to the following document:

D1: WO 98/20073 (cf. claims 1, 28, 37, 38)

Hydrophobic polymers incompatible with starch containing , as filler, a starch complex dispersed in the hydrophobic polymeric matrix, bound to the polymer matrix by means of coupling agents containing groups compatible with the matrix and with the starch complex, have already been disclosed in D1. The starch complex of D1 is in the form of particles with a numeral average size of less than 1 μm .

Even though D1 does not explicitly mention the solubility in water and the second-derivative IR absorption, it is reasonable to assume that the starch complex quoted therein shows the same IR absorption and water-solubility as those indicated in the present application, since the composition of D1 has been prepared in the same way as the present composition.

Therefore, the subject-matter of the present claim 1 is not novel under article 33(2) PCT.

The subject-matter of claims 2 to 24 is either not novel with respect to D1 or would not appear to be inventive with regard to this teaching (Art. 33 (2) and (3) PCT).

PCT

REQUEST

The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty.

For receiving Office use only

PCT/EP 99 / 07038

International Application No.

22 SEP 1999

(22.09.1999)

International Filing Date

EUROPEAN PATENT OFFICE
PCT INTERNATIONAL APPLICATION

Name of receiving Office and "PCT International Application"

Applicant's or agent's file reference

(if desired) (12 characters maximum) PC273PR

Box No. I TITLE OF INVENTION

"Polymers of a hydrophobic nature, filled with starch complexes"

Box No. II APPLICANT

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

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IT

☐ This person is also inventor.

Telephone No.

Facsimile No.

Teleprinter No.

State (that is, country) of nationality:

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State (that is, country) of residence:

IT

This person is applicant for the purposes of:

☐ all designated States

☒ all designated States except the United States of America

☐ the United States of America only

☐ the States indicated in the Supplemental Box

Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

BASTIOLI, Catia
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This person is:

☐ applicant only

☒ applicant and inventor

☐ inventor only (If this check-box is marked, do not fill in below.)

State (that is, country) of nationality:

IT

State (that is, country) of residence:

IT

This person is applicant for the purposes of:

☐ all designated States

☐ all designated States except the United States of America

☒ the United States of America only

☐ the States indicated in the Supplemental Box

☒ Further applicants and/or (further) inventors are indicated on a continuation sheet.

Box No. IV AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE

The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as:

☒ agent

☐ common representative

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

RAMBELLI, Paolo (IT); JACOBACCI, Filippo (IT); JACOBACCI, Guido (IT); SACONNEY, Piero (IT); QUINTERNO, Giuseppe (IT); GERBINO, Angelo (IT); SERRA, Francesco (IT); FIORAVANTI, Corrado (IT); all c/o JACOBACCI & PERANI S.p.A., Corso Regio Parco 27, I-10152 TORINO (Italy)

Telephone No.

(39) (011) 2440311

Facsimile No.

(39) (011) 286300 / 286676

Teleprinter No.

☐ Address for correspondence: Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.

Zur Kass
(A) 010
4713

Continuation of Box No. III FURTHER APPLICANTS AND/OR (FURTHER) INVENTORS

If none of the following sub-boxes is used, this sheet should not be included in the request.

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

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Via Mora e Gibin 9
I-28010 FONTANETO D'AGOGNA (Novara)
IT^Δ

This person is:

☐ applicant only☒ applicant and inventor☐ inventor only (If this check-box is marked, do not fill in below.)

State (that is, country) of nationality:

IT

State (that is, country) of residence:

IT

This person is applicant for the purposes of:

☐ all designated States☐ all designated States except the United States of America☒ the United States of America only☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

MONTINO, Alessandro
Via Bellotti 15
I-27038 ROBBIO LOMELLINA (Pavia)
IT^Δ

This person is:

☐ applicant only☒ applicant and inventor☐ inventor only (If this check-box is marked, do not fill in below.)

State (that is, country) of nationality:

IT

State (that is, country) of residence:

IT

This person is applicant for the purposes of:

☐ all designated States☐ all designated States except the United States of America☒ the United States of America only☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

This person is:

☐ applicant only☐ applicant and inventor☐ inventor only (If this check-box is marked, do not fill in below.)

State (that is, country) of nationality:

State (that is, country) of residence:

This person is applicant for the purposes of:

☐ all designated States☐ all designated States except the United States of America☐ the United States of America only☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

This person is:

☐ applicant only☐ applicant and inventor☐ inventor only (If this check-box is marked, do not fill in below.)

State (that is, country) of nationality:

State (that is, country) of residence:

This person is applicant for the purposes of:

☐ all designated States☐ all designated States except the United States of America☐ the United States of America only☐ the States indicated in the Supplemental Box☐ Further applicants and/or (further) inventors are indicated on another continuation sheet.

Box No.V DESIGNATION OF STATES

The following designations are hereby made under Rule 4.9(a) (mark the applicable check-boxes; at least one must be marked):

Regional Patent

- ☒ **AP ARIPO Patent:** GH Ghana, GM Gambia, KE Kenya, LS Lesotho, MW Malawi, SD Sudan, SL Sierra Leone, SZ Swaziland, UG Uganda, ZW Zimbabwe, and any other State which is a Contracting State of the Harare Protocol and of the PCT
- ☒ **EA Eurasian Patent:** AM Armenia, AZ Azerbaijan, BY Belarus, KG Kyrgyzstan, KZ Kazakhstan, MD Republic of Moldova, RU Russian Federation, TJ Tajikistan, TM Turkmenistan, and any other State which is a Contracting State of the Eurasian Patent Convention and of the PCT
- ☒ **EP European Patent:** AT Austria, BE Belgium, CH and LI Switzerland and Liechtenstein, CY Cyprus, DE Germany, DK Denmark, ES Spain, FI Finland, FR France, GB United Kingdom, GR Greece, IE Ireland, IT Italy, LU Luxembourg, MC Monaco, NL Netherlands, PT Portugal, SE Sweden, and any other State which is a Contracting State of the European Patent Convention and of the PCT
- ☒ **OA OAPI Patent:** BF Burkina Faso, BJ Benin, CF Central African Republic, CG Congo, CI Côte d'Ivoire, CM Cameroon, GA Gabon, GN Guinea, GW Guinea-Bissau, ML Mali, MR Mauritania, NE Niger, SN Senegal, TD Chad, TG Togo, and any other State which is a member State of OAPI and a Contracting State of the PCT (if other kind of protection or treatment desired, specify on dotted line)

National Patent (if other kind of protection or treatment desired, specify on dotted line):

- | | |
|--|--|
| <input checked="" type="checkbox"/> AE United Arab Emirates | <input checked="" type="checkbox"/> LR Liberia |
| <input checked="" type="checkbox"/> AL Albania | <input checked="" type="checkbox"/> LS Lesotho |
| <input checked="" type="checkbox"/> AM Armenia | <input checked="" type="checkbox"/> LT Lithuania |
| <input checked="" type="checkbox"/> AT Austria | <input checked="" type="checkbox"/> LU Luxembourg |
| <input checked="" type="checkbox"/> AU Australia | <input checked="" type="checkbox"/> LV Latvia |
| <input checked="" type="checkbox"/> AZ Azerbaijan | <input checked="" type="checkbox"/> MD Republic of Moldova |
| <input checked="" type="checkbox"/> BA Bosnia and Herzegovina | <input checked="" type="checkbox"/> MG Madagascar |
| <input checked="" type="checkbox"/> BB Barbados | <input checked="" type="checkbox"/> MK The former Yugoslav Republic of Macedonia |
| <input checked="" type="checkbox"/> BG Bulgaria | |
| <input checked="" type="checkbox"/> BR Brazil | <input checked="" type="checkbox"/> MN Mongolia |
| <input checked="" type="checkbox"/> BY Belarus | <input checked="" type="checkbox"/> MW Malawi |
| <input checked="" type="checkbox"/> CA Canada | <input checked="" type="checkbox"/> MX Mexico |
| <input checked="" type="checkbox"/> CH and LI Switzerland and Liechtenstein | <input checked="" type="checkbox"/> NO Norway |
| <input checked="" type="checkbox"/> CN China | <input checked="" type="checkbox"/> NZ New Zealand |
| <input checked="" type="checkbox"/> CU Cuba | <input checked="" type="checkbox"/> PL Poland |
| <input checked="" type="checkbox"/> CZ Czech Republic | <input checked="" type="checkbox"/> PT Portugal |
| <input checked="" type="checkbox"/> DE Germany | <input checked="" type="checkbox"/> RO Romania |
| <input checked="" type="checkbox"/> DK Denmark | <input checked="" type="checkbox"/> RU Russian Federation |
| <input checked="" type="checkbox"/> EE Estonia | <input checked="" type="checkbox"/> SD Sudan |
| <input checked="" type="checkbox"/> ES Spain | <input checked="" type="checkbox"/> SE Sweden |
| <input checked="" type="checkbox"/> FI Finland | <input checked="" type="checkbox"/> SG Singapore |
| <input checked="" type="checkbox"/> GB United Kingdom | <input checked="" type="checkbox"/> SI Slovenia |
| <input checked="" type="checkbox"/> GD Grenada | <input checked="" type="checkbox"/> SK Slovakia |
| <input checked="" type="checkbox"/> GE Georgia | <input checked="" type="checkbox"/> SL Sierra Leone |
| <input checked="" type="checkbox"/> GH Ghana | <input checked="" type="checkbox"/> TJ Tajikistan |
| <input checked="" type="checkbox"/> GM Gambia | <input checked="" type="checkbox"/> TM Turkmenistan |
| <input checked="" type="checkbox"/> HR Croatia | <input checked="" type="checkbox"/> TR Turkey |
| <input checked="" type="checkbox"/> HU Hungary | <input checked="" type="checkbox"/> TT Trinidad and Tobago |
| <input checked="" type="checkbox"/> ID Indonesia | <input checked="" type="checkbox"/> UA Ukraine |
| <input checked="" type="checkbox"/> IL Israel | <input checked="" type="checkbox"/> UG Uganda |
| <input checked="" type="checkbox"/> IN India | <input checked="" type="checkbox"/> US United States of America |
| <input checked="" type="checkbox"/> IS Iceland | |
| <input checked="" type="checkbox"/> JP Japan | <input checked="" type="checkbox"/> UZ Uzbekistan |
| <input checked="" type="checkbox"/> KE Kenya | <input checked="" type="checkbox"/> VN Viet Nam |
| <input checked="" type="checkbox"/> KG Kyrgyzstan | <input checked="" type="checkbox"/> YU Yugoslavia |
| <input checked="" type="checkbox"/> KP Democratic People's Republic of Korea | <input checked="" type="checkbox"/> ZA South Africa |
| | <input checked="" type="checkbox"/> ZW Zimbabwe |
| <input checked="" type="checkbox"/> KR Republic of Korea | Check-boxes reserved for designating States which have become party to the PCT after issuance of this sheet: |
| <input checked="" type="checkbox"/> KZ Kazakhstan | <input checked="" type="checkbox"/> CR COSTA RICA |
| <input checked="" type="checkbox"/> LC Saint Lucia | <input checked="" type="checkbox"/> DM DOMINICA |
| <input checked="" type="checkbox"/> LK Sri Lanka | <input checked="" type="checkbox"/> TZ TANZANIA |

Precautionary Designation Statement: In addition to the designations made above, the applicant also makes under Rule 4.9(b) all other designations which would be permitted under the PCT except any designation(s) indicated in the Supplemental Box as being excluded from the scope of this statement. The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit. (Confirmation of a designation consists of the filing of a notice specifying that designation and the payment of the designation and confirmation fees. Confirmation must reach the receiving Office within the 15-month time limit.)



Box No. VI PRIORITY CLAIM		<input type="checkbox"/> Further priority claims are indicated in the Supplemental Box.		
Filing date of earlier application (day/month/year)	Number of earlier application	Where earlier application is:		
		national application: country	regional application:* regional Office	international application: receiving Office
item (1) (22.09.1998) 22 SEPTEMBER 1998	TO98A000800	ITALY		
item (2)				
item (3)				

☐ The receiving Office is requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) (only if the earlier application was filed with the Office which for the purposes of the present international application is the receiving Office) identified above as item(s):

* Where the earlier application is an ARIPO application, it is mandatory to indicate in the Supplemental Box at least one country party to the Paris Convention for the Protection of Industrial Property for which that earlier application was filed (Rule 4.10(b)(iii)). See Supplemental Box.

Box No. VII INTERNATIONAL SEARCHING AUTHORITY

Choice of International Searching Authority (ISA)
(if two or more International Searching Authorities are competent to carry out the international search, indicate the Authority chosen; the two-letter code may be used):

ISA / EP

Request to use results of earlier search; reference to that search (if an earlier search has been carried out by or requested from the International Searching Authority):

Date (day/month/year)

Number

Country (or regional Office)

Box No. VIII CHECK LIST; LANGUAGE OF FILING

This international application contains the following number of sheets:

request : 4

description (excluding sequence listing part) : 18

claims : 5

abstract : 1

drawings :

sequence listing part of description :

Total number of sheets : 28

This international application is accompanied by the item(s) marked below:

1. ☒ fee calculation sheet
2. ☒ separate signed power of attorney (to follows)
3. ☐ copy of general power of attorney; reference number, if any:
4. ☐ statement explaining lack of signature
5. ☒ priority document(s) identified in Box No. VI as item(s): (1) (to follow)
6. ☐ translation of international application into (language):
7. ☐ separate indications concerning deposited microorganism or other biological material
8. ☐ nucleotide and/or amino acid sequence listing in computer readable form
9. ☐ other (specify):

Figure of the drawings which should accompany the abstract:

Language of filing of the international application: English

Box No. IX SIGNATURE OF APPLICANT OR AGENT

Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the request).


RAMBELLI, Paolo

For receiving Office use only		2. Drawings: <input type="checkbox"/> received: <input type="checkbox"/> not received:
1. Date of actual receipt of the purported international application:	22 SEP 1999 (22.09.1999)	
3. Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application:		
4. Date of timely receipt of the required corrections under PCT Article 11(2):		
5. International Searching Authority (if two or more are competent): ISA /	6. <input type="checkbox"/> Transmittal of search copy delayed until search fee is paid.	

For International Bureau use only
Date of receipt of the record copy by the International Bureau:



The demand must be filed with the competent International Preliminary Examining Authority or, if two or more Authorities are competent, with the one chosen by the applicant. The full name or two-letter code of that Authority may be indicated by the applicant on the line below:
IPEA/ _____

PCT

CHAPTER II

DEMAND

under Article 31 of the Patent Cooperation Treaty:
The undersigned requests that the international application specified below be the subject of international preliminary examination according to the Patent Cooperation Treaty and hereby elects all eligible States (except where otherwise indicated).

For International Preliminary Examining Authority use only	
Identification of IPEA	Date of receipt of DEMAND
Box No. I IDENTIFICATION OF THE INTERNATIONAL APPLICATION	
Applicant's or agent's file reference PC273PR	
International application No. PCT/EP99/07038	International filing date (day/month/year) 22 September 1999 (22.09.1999)
(Earliest) Priority date (day/month/year) 22 September 1998 (22.09.1998)	
Title of invention "Polymers of a hydrophobic nature, filed with starch complexes"	
Box No. II APPLICANT(S)	
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.) NOVAMONT S.p.A. Via Fauser 8 I-28100 NOVARA (Italy)	
Telephone No.:	
Facsimile No.:	
Teleprinter No.:	
State (that is, country) of nationality: IT	State (that is, country) of residence: IT
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.) BASTIOLI, Catia Via della Noce 63 I-28100 NOVARA (Italy)	
State (that is, country) of nationality: IT	State (that is, country) of residence: IT
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.) BELLOTTI, Vittorio Via Mora e Gibin 9 I-28010 FONTANETO D'AGOGNA (Novara) Italy	
State (that is, country) of nationality: IT	State (that is, country) of residence: IT
<input checked="" type="checkbox"/> Further applicants are indicated on a continuation sheet.	



Sheet No: 2

International application No.
PCT/EP99/07038

Continuation of Box No. II APPLICANT(S)	
<i>If none of the following sub-boxes is used, this sheet should not be included in the demand.</i>	
<p>Name and address: <small>(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)</small></p> <p>MONTINO, Alessandro Via Bellotti 15 I-27038 ROBBIO LOMELLINA (Pavia) Italy</p>	
<p>State <small>(that is, country)</small> of nationality:</p> <p style="text-align: center;">IT</p>	<p>State <small>(that is, country)</small> of residence:</p> <p style="text-align: center;">IT</p>
<p>Name and address: <small>(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)</small></p>	
<p>State <small>(that is, country)</small> of nationality:</p>	<p>State <small>(that is, country)</small> of residence:</p>
<p>Name and address: <small>(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)</small></p>	
<p>State <small>(that is, country)</small> of nationality:</p>	<p>State <small>(that is, country)</small> of residence:</p>
<p>Name and address: <small>(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)</small></p>	
<p>State <small>(that is, country)</small> of nationality:</p>	<p>State <small>(that is, country)</small> of residence:</p>
<p>Name and address: <small>(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)</small></p>	
<p>State <small>(that is, country)</small> of nationality:</p>	<p>State <small>(that is, country)</small> of residence:</p>
<p><input type="checkbox"/> Further applicants are indicated on another continuation sheet.</p>	

Box No. III AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCEThe following person is ☒ agent ☐ common representativeand ☒ has been appointed earlier and represents the applicant(s) also for international preliminary examination.☐ is hereby appointed and any earlier appointment of (an) agent(s)/common representative is hereby revoked.☐ is hereby appointed, specifically for the procedure before the International Preliminary Examining Authority, in addition to the agent(s)/common representative appointed earlier.Name and address: *(Family name followed by given name; for a legal entity, full official designation.
The address must include postal code and name of country.)*

RAMBELLI, Paolo (IT)

c/o JACOBACCI & PERANI S.p.A.
Corso Regio Parco 27
I-10152 TORINO (Italy)

Telephone No.:

+39 +011 2440311

Facsimile No.:

+39 +011 286300 / 286676

Teleprinter No.:

☐ Address for correspondence: Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.**Box No. IV BASIS FOR INTERNATIONAL PRELIMINARY EXAMINATION****Statement concerning amendments:***

1. The applicant wishes the international preliminary examination to start on the basis of:

☒ the international application as originally filedthe description ☐ as originally filed☐ as amended under Article 34the claims ☐ as originally filed☐ as amended under Article 19 (together with any accompanying statement)☐ as amended under Article 34the drawings ☐ as originally filed☐ as amended under Article 342. ☐ The applicant wishes any amendment to the claims under Article 19 to be considered as reversed.3. ☐ The applicant wishes the start of the international preliminary examination to be postponed until the expiration of 20 months from the priority date unless the International Preliminary Examining Authority receives a copy of any amendments made under Article 19 or a notice from the applicant that he does not wish to make such amendments (Rule 69.1(d)). *(This check-box may be marked only where the time limit under Article 19 has not yet expired.)*

* Where no check-box is marked, international preliminary examination will start on the basis of the international application as originally filed or, where a copy of amendments to the claims under Article 19 and/or amendments of the international application under Article 34 are received by the International Preliminary Examining Authority before it has begun to draw up a written opinion or the international preliminary examination report, as so amended.

Language for the purposes of international preliminary examination: ENGLISH☒ which is the language in which the international application was filed.☐ which is the language of a translation furnished for the purposes of international search.☐ which is the language of publication of the international application.☐ which is the language of the translation (to be) furnished for the purposes of international preliminary examination.**Box No. V ELECTION OF STATES**The applicant hereby elects all eligible States *(that is, all States which have been designated and which are bound by Chapter II of the PCT)*

excluding the following States which the applicant wishes not to elect:



Box No. VI CHECK LIST

The demand is accompanied by the following elements, in the language referred to in Box No. IV, for the purposes of international preliminary examination:

- | | | |
|--|---|--------|
| 1. translation of international application | : | sheets |
| 2. amendments under Article 34 | : | sheets |
| 3. copy (or, where required, translation) of amendments under Article 19 | : | sheets |
| 4. copy (or, where required, translation) of statement under Article 19 | : | sheets |
| 5. letter | : | sheets |
| 6. other (<i>specify</i>) | : | sheets |

For International Preliminary
Examining Authority use only

received not received


<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>

The demand is also accompanied by the item(s) marked below:

- | | |
|--|---|
| 1. <input checked="" type="checkbox"/> fee calculation sheet | 4. <input type="checkbox"/> statement explaining lack of signature |
| 2. <input type="checkbox"/> separate signed power of attorney | 5. <input type="checkbox"/> nucleotide and or amino acid sequence listing in computer readable form |
| 3. <input type="checkbox"/> copy of general power of attorney; reference number, if any: | 6. <input type="checkbox"/> other (<i>specify</i>): |

Box No. VII SIGNATURE OF APPLICANT, AGENT OR COMMON REPRESENTATIVE

Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the demand).


RAMBELLI, Paolo

For International Preliminary Examining Authority use only

1. Date of actual receipt of DEMAND:

2. Adjusted date of receipt of demand due to CORRECTIONS under Rule 60.1(b):

3. ☐ The date of receipt of the demand is AFTER the expiration of 19 months from the priority date and item 4 or 5, below, does not apply.

☐ The applicant has been informed accordingly.

4. ☐ The date of receipt of the demand is WITHIN the period of 19 months from the priority date as extended by virtue of Rule 80.5.

5. ☐ Although the date of receipt of the demand is after the expiration of 19 months from the priority date, the delay in arrival is EXCUSED pursuant to Rule 82.

For International Bureau use only

Demand received from IPEA on:

PATENT COOPERATION TREATY

Bee

From the INTERNATIONAL BUREAU

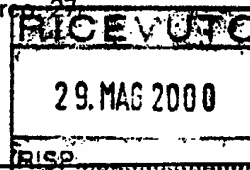
PCT

INFORMATION CONCERNING ELECTED
OFFICES NOTIFIED OF THEIR ELECTION

(PCT Rule 61.3)

To:

RAMBELLI, Paolo
Jacobacci & Perani S.p.A.
Corso Regio Parco, 27
I-10152 Torino
ITALIE



Date of mailing (day/month/year) 15 May 2000 (15.05.00)		
Applicant's or agent's file reference PC273PR		
IMPORTANT INFORMATION		
International application No. PCT/EP99/07038 ✓	International filing date (day/month/year) 22 September 1999 (22.09.99)	Priority date (day/month/year) 22 September 1998 (22.09.98)
Applicant NOVAMONT S.P.A. et al ✓		

1. The applicant is hereby informed that the International Bureau has, according to Article 31(7), notified each of the following Offices of its election:

AP : GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW ✓
 EP : AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE
 National : AU, BG, BR, CA, CN, CZ, DE, IL, JP, KP, KR, MN, NO, NZ, PL, RO, RU, SE, SK, US

2. The following Offices have waived the requirement for the notification of their election; the notification will be sent to them by the International Bureau only upon their request:

EA : AM, AZ, BY, KG, KZ, MD, RU, TJ, TM ✓
 OA : BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG
 National : AE, AL, AM, AT, AZ, BA, BB, BY, CH, CR, CU, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM,
 HR, HU, ID, IN, IS, KE, KG, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MW, MX, PT, SD, SG, SI,
 SL, TJ, TM, TR, TT, TZ, UA, UG, UZ, VN, YU, ZA, ZW

3. The applicant is reminded that he must enter the "national phase" before the expiration of 30 months from the priority date before each of the Offices listed above. This must be done by paying the national fee(s) and furnishing, if prescribed, a translation of the international application (Article 39(1)(a)), as well as, where applicable, by furnishing a translation of any annexes of the international preliminary examination report (Article 36(3)(b) and Rule 74.1).

Some offices have fixed time limits expiring later than the above-mentioned time limit. For detailed information about the applicable time limits and the acts to be performed upon entry into the national phase before a particular Office, see Volume II of the PCT Applicant's Guide.

The entry into the European regional phase is postponed until 31 months from the priority date for all States designated for the purposes of obtaining a European patent.

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No. (41-22) 740.14.35	Authorized officer: R. E. Stoffel Telephone No. (41-22) 338.83.38
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PATENT COOPERATION TREATY

PCT

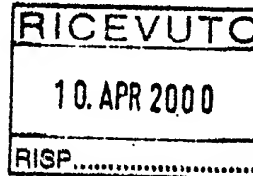
From the INTERNATIONAL BUREAU

NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

To:

RAMBELLI, Paolo
Jacobacci & Perani S.p.A.
Corso Regio Parco, 27
I-10152 Torino
ITALIE



Date of mailing (day/month/year) 30 March 2000 (30.03.00)		IMPORTANT NOTICE	
Applicant's or agent's file reference PC273PR			
International application No. PCT/EP99/07038 ✓	International filing date (day/month/year) 22 September 1999 (22.09.99)	Priority date (day/month/year) 22 September 1998 (22.09.98) ✓	
Applicant NOVAMONT S.P.A. et al ✓			

1. Notice is hereby given that the International Bureau has communicated, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this Notice:
AU,CN,JP,KP,KR,US ✓

In accordance with Rule 47.1(c), third sentence, those Offices will accept the present Notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:
**AE,AL,AM,AP,AT,AZ,BA,BB,BG,BR,BY,CA,CH,CR,CU,CZ,DE,DK,DM,EA,EE,EP,ES,FI,GB,GD,GE, ✓
GH,GM,HR,HU,ID,IL,IN,IS,KE,KG,KZ,LC,LK,LR,LS,LT,LU,LV,MD,MG,MK,MN,MW,MX,NO,NZ,OA,
PL,PT,RO,RU,SD,SE,SG,SI,SK,SL,TJ,TM,TR,TT,TZ,UA,UG,UZ,VN,YU,ZA,ZW**
The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).

3. Enclosed with this Notice is a copy of the international application as published by the International Bureau on
30 March 2000 (30.03.00) under No. WO 00/17270

REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, a demand for international preliminary examination must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))

If the applicant wishes to proceed with the international application in the national phase, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and Volume II of the PCT Applicant's Guide.

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer J. Zahra
Facsimile No. (41-22) 740.14.35	Telephone No. (41-22) 338.83.38



Continuation of Form PCT/IB/308

NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF
THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES

Date of mailing (day/month/year) 30 March 2000 (30.03.00)	IMPORTANT NOTICE
Applicant's or agent's file reference PC273PR	International application No. PCT/EP99/07038
<p>The applicant is hereby notified that, at the time of establishment of this Notice, the time limit under Rule 46.1 for making amendments under Article 19 has not yet expired and the International Bureau had received neither such amendments nor a declaration that the applicant does not wish to make amendments.</p>	



PATENT COOPERATION TREATY

Bee 28

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

RAMBELLI, Paolo
JACOBACCI & PERANI S.P.A.
Corso Regio Parco, 27
10152 Torino
ITALIE

RICEVUTO

29. DIC 2000

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NOTIFICATION OF TRANSMITTAL OF
THE INTERNATIONAL PRELIMINARY
EXAMINATION REPORT

(PCT Rule 71.1)

Date of mailing
(day/month/year)

21. 12. 00

Applicant's or agent's file reference
PC273PR

IMPORTANT NOTIFICATION

International application No.
PCT/EP99/07038 ✓

International filing date (day/month/year)
22/09/1999 ✓

Priority date (day/month/year)
22/09/1998 ✓

Applicant
NOVAMONT S.P.A. et al. ✓

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/



European Patent Office
D-80298 Munich
Tel. +49 89 2399 - 0 Tx: 523656 epmu d
Fax: +49 89 2399 - 4465

Authorized officer

Le Bolloch, C

Tel. +49 89 2399-8091





PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)



Applicant's or agent's file reference PC273PR	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/EP99/07038 ✓	International filing date (day/month/year) 22/09/1999 ✓	Priority date (day/month/year) 22/09/1998 ✓
International Patent Classification (IPC) or national classification and IPC C08L29/04		
Applicant NOVAMONT S.P.A. et al. ✓		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 4 sheets, including this cover sheet.

☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

 These annexes consist of a total of 5 sheets.

3. This report contains indications relating to the following items:
 - I ☒ Basis of the report
 - II ☐ Priority
 - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
 - IV ☐ Lack of unity of invention
 - V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
 - VI ☐ Certain documents cited
 - VII ☐ Certain defects in the international application
 - VIII ☐ Certain observations on the international application

Date of submission of the demand 18/04/2000	Date of completion of this report 21. 12. 00
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Boletti, C Telephone No. +49 89 2399 8527 



**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/EP99/07038

I. Basis of the report

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).)*:

Description, pages:

1,2,4-18	as originally filed	✓
3	with telefax of	19/10/2000 ✓

Claims, No.:

1-24	with telefax of	19/10/2000 ✓
------	-----------------	--------------

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
 - ☐ the language of publication of the international application (under Rule 48.3(b)).
 - ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).
3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:
- ☐ contained in the international application in written form.
 - ☐ filed together with the international application in computer readable form.
 - ☐ furnished subsequently to this Authority in written form.
 - ☐ furnished subsequently to this Authority in computer readable form.
 - ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
 - ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:



**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/EP99/07038

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims
	No: Claims 1
Inventive step (IS)	Yes: Claims
	No: Claims 2-24
Industrial applicability (IA)	Yes: Claims 1-24
	No: Claims

- 2. Citations and explanations**
see separate sheet



ITEM V

Reference is made to the following document:

D1: WO 98/20073 (cf. claims 1, 28, 37, 38)

Hydrophobic polymers incompatible with starch containing , as filler, a starch complex dispersed in the hydrophobic polymeric matrix, bound to the polymer matrix by means of coupling agents containing groups compatible with the matrix and with the starch complex, have already been disclosed in D1. The starch complex of D1 is in the form of particles with a numeral average size of less than 1 μm .

Even though D1 does not explicitly mention the solubility in water and the second-derivative IR absorption, it is reasonable to assume that the starch complex quoted therein shows the same IR absorption and water-solubility as those indicated in the present application, since the composition of D1 has been prepared in the same way as the present composition.

Therefore, the subject-matter of the present claim 1 is not novel under article 33(2) PCT.

The subject-matter of claims 2 to 24 is either not novel with respect to D1 or would not appear to be inventive with regard to this teaching (Art. 33 (2) and (3) PCT).



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not identified further, and which can act as a compatibilizing agent between starch and rubber.

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It has ~~now~~ unexpectedly been found that it is possible to disperse, in hydrophobic polymers incompatible with starch, starch complexes characterized by second-derivative FTIR absorption in the region of $940-950\text{ cm}^{-1}$ or by XR diffraction peaks in the regions of $11^{\circ}-13^{\circ}$ and $19^{\circ}-21^{\circ}$ of 2θ , in the form of particles with poor solubility in water and having numeral average size of less ~~than 3 microns~~, preferably less than 1 micron, and which are fixed to the polymer matrix by means of (a) coupling agent(s) containing groups which can interact with the polymer matrix and with the complex (external coupling agent) or by means of reactive groups present in the complex capable of being fixed to the polymeric matrix thus acting as internal coupling agent. In this case, the use of the external coupling agent can be omitted.

This is, for example, the case of matrices comprising a biodegradable polymer such as the aliphatic or aliphatic-aromatic polyesters, the aliphatic polyamides, polyamides-polyesters, polyurethane-polyesters and the like.

As it will be specified hereinafter, the complex usable with the biodegradable matrices of the type above indicated, is a complex of starch with a ^{complexing agent} ~~polymer~~ different from the polymer forming the polymeric matrix and from the ethylene-vinylalcohol copolymers, ~~or with other complexing agents.~~

< The present invention provides hydrophobic polymers filled with starch complexes and a process for their preparation, as defined in the appended claims >



CLAIMS

1. Hydrophobic polymers incompatible with starch containing, as a filler, a starch complex in the form of particles dispersed in the hydrophobic polymeric matrix and bound to the polymer matrix by means of coupling agents containing groups compatible with the matrix and with the starch complex or by means of reactive groups present in the starch-complex capable of being fixed to the polymeric matrix, characterised in that the starch complex is in the form of particles with a numeral average size of less than 1 micron, it has a solubility in water at 100°C of less than 20% and is characterised by a second-derivative IR absorption in the region of 940-950 cm^{-1} , and wherein the starch complex, in the case where the hydrophobic biodegradable polymers of the matrix are selected from the group consisting of the aliphatic or aliphatic-aromatic polyesters, the aliphatic polyamides, polyamides-polyesters, polyurethane polyesters, polyurethane-polyamides, polyurea-polyesters, is a complex of starch with a complexing agent different from the polymer forming the matrix and from ethylene-vinylalcohol copolymers.

2. Polymers according to claim 1, wherein the coupling agent is selected from the groups consisting of a vinyl silane, an alkyl titanate, and bis-3-triethoxysilylpropyl tetrasulphide.

3. Polymers according to claims 1 or 2, wherein the complexing agent different from the polymer forming the polymeric matrix is selected from the group consisting of polylactic acid, polyglycolic acid, poly(lactic-glycolic) acid copolymers, ethylene-acrylic acid copolymers, ethylene-vinylacetate copolymers.

4. Polymers according to any of claims 1 to 3, in which the quantity of filler comprising the complex dispersed in



the hydrophobic polymer is from 0.5 to 50% by weight.

5. Polymers according to any of claims 1 to 4, in which the starch complexes are produced from compositions of starch with polymers compatible with starch containing lyophilic groups and lyophobic sequences, wherein the starch complex is present and from which a micro-dispersion of particles with numeral average diameters of less than 1 micron is formed by treatment in water at 100°C under stirring.
6. Polymers according to claims 1 to 5, produced with the use of compositions having a water content of less than 20%, and higher than 2% by weight, and a Tg below 0°C.
7. Polymers according to claim 5, in which the polymer which can form complexes with starch is selected from the group comprising copolymers of ethylene with polar monomers.
8. Polymers according to claim 7, in which the copolymer is selected from the group comprising copolymers of ethylene with vinyl alcohol, vinyl acetate and acrylic acid.
9. Polymers according to claim 8, in which the ethylene/vinyl alcohol copolymer contains from 50 to 75% of vinyl alcohol in moles.
10. Polymers according to claim 5, in which the polymer which can complex with the starch is selected from copolymers of polyester/polyurethane, polyamide/polyesters, aliphatic and aliphatic aromatic polyesters and polyamides.
11. Polymers according to claims 1 or 2, wherein the starch complexing agent is a fatty acid or a derivative thereof.
12. Polymers according to claims 1 or 2, wherein the starch complexing agent contains reactive groups for the hydrophobic matrix.



13. Polymers according to any of claim 1 to 12, in which the hydrophobic polymer incompatible with starch is selected from the group consisting of ethylene polymers and copolymers, crystalline propylene polymers and copolymers, aromatic polyester resins, polyamides, polyoxymethylene resins, polyphenylene oxide resins, and polycarbonates.

14. Polymers according to any of claims 1 to 12, in which the hydrophobic polymer is a rubber selected from the group consisting of styrene-butadiene rubbers, polybutadiene rubbers, polyisoprene rubbers, ethylene-propylene and ethylene-propylene-diene rubbers, and natural rubber.

15. A method for preparing filled polymers according to any of claims 1 to 14, in which a composition comprising the starch/polymer complex, forming part of a continuous interpenetrated structure between the complexing polymer and the complex is mixed, in the melt state or under hot mastication conditions, with the hydrophobic polymer incompatible with starch, in the presence of coupling agents containing groups reactive with the polymer matrix and with the complex.

16. A method preparing filled polymers according to any of claims 1 to 14, in which a composition comprising the starch/polymer complex is mixed with a rubber at a processing temperature between 140 and 160°C, in the presence of coupling agents containing groups reactive with the polymer matrix and with the complex.

17. A method according to claims 15 and 16, in which the coupling agent is selected from vinyl and tetrasulphide silanes and alkyl titanates.

18. A method according to any of claims 15, 16 and 17 in which the coupling agent is used in a quantity of from 0.5 to 10% by weight of the complex.



19. A method for preparing filled polymers according to claims 1 to 14, wherein the polymeric matrix is a biodegradable polymer selected from the group consisting of the aliphatic-aromatic polyesters, the aliphatic polyamides, the polyamides-polyesters, polyurethane-polyesters, polyurethane-polyamides and polyurea-polyesters comprising melt-mixing the polymer forming the polymeric matrix with a complex of starch having the characteristics as set forth in claim 1 and further characterised by being formed of starch complexed with a complexing agent different from the polymer forming the matrix and from the ethylene-vinylalcohol copolymers.

20. A method according to claim 19, wherein the starch complex is preformed or formed during melt-mixing.

21. Shaped articles obtainable from the hydrophobic polymers of claims 1 to 14.

22. Shaped articles obtainable from the hydrophobic polymers of claims 1 to 14, wherein the hydrophobic polymer is selected from the group consisting of the aliphatic and aliphatic-aromatic polyesters, polyurethane-polyamides, polyurea-polyesters, and polyurethane-polyesters.

23. Films and compostable bags obtainable from the hydrophobic polymers of claims 1 to 14.

24. Tyres obtainable from the rubbers of claim 14.



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This is, for example, the case of matrices comprising a biodegradable polymer such as the aliphatic or aliphatic-aromatic polyesters, the aliphatic polyamides, polyamides-polyesters, polyurethane-polyesters and the like.

As it will be specified hereinafter, the complex usable with the biodegradable matrices of the type above indicated, is a complex of starch with a ^{complexing agent} polymer different from the polymer forming the polymeric matrix and from the ethylene-vinylalcohol copolymers, ~~or with other complexing agents~~.

< The present invention provides hydrophobic polymers filled with starch complexes and a process for their preparation, as defined in the appended claims >



CLAIMS

1. Hydrophobic polymers incompatible with starch containing, as a filler, a starch complex in the form of particles dispersed in the hydrophobic polymeric matrix and bound to the polymer matrix by means of coupling agents containing groups compatible with the matrix and with the starch complex or by means of reactive groups present in the starch-complex capable of being fixed to the polymeric matrix, characterised in that the starch complex is in the form of particles with a numeral average size of less than 1 micron, it has a solubility in water at 100°C of less than 20% and is characterised by a second-derivative IR absorption in the region of 940-950 cm^{-1} , and wherein the starch complex, in the case where the hydrophobic biodegradable polymers of the matrix are selected from the group consisting of the aliphatic or aliphatic-aromatic polyesters, the aliphatic polyamides, polyamides-polyesters, polyurethane polyesters, polyurethane-polyamides, polyurea-polyesters, is a complex of starch with a complexing agent different from the polymer forming the matrix and from ethylene-vinylalcohol copolymers.

2. Polymers according to claim 1, wherein the coupling agent is selected from the groups consisting of a vinyl silane, an alkyl titanate, and bis-3-triethoxysilylpropyl tetrasulphide.

3. Polymers according to claims 1 or 2, wherein the complexing agent different from the polymer forming the polymeric matrix is selected from the group consisting of polylactic acid, polyglycolic acid, poly(lactic-glycolic) acid copolymers, ethylene-acrylic acid copolymers, ethylene-vinylacetate copolymers.

4. Polymers according to any of claims 1 to 3, in which the quantity of filler comprising the complex dispersed in



the hydrophobic polymer is from 0.5 to 50% by weight.

5. Polymers according to any of claims 1 to 4, in which the starch complexes are produced from compositions of starch with polymers compatible with starch containing lyophilic groups and lyophobic sequences, wherein the starch complex is present and from which a micro-dispersion of particles with numeral average diameters of less than 1 micron is formed by treatment in water at 100°C under stirring.

6. Polymers according to claims 1 to 5, produced with the use of compositions having a water content of less than 20%, and higher than 2% by weight, and a Tg below 0°C.

7. Polymers according to claim 5, in which the polymer which can form complexes with starch is selected from the group comprising copolymers of ethylene with polar monomers.

8. Polymers according to claim 7, in which the copolymer is selected from the group comprising copolymers of ethylene with vinyl alcohol, vinyl acetate and acrylic acid.

9. Polymers according to claim 8, in which the ethylene/vinyl alcohol copolymer contains from 50 to 75% of vinyl alcohol in moles.

10. Polymers according to claim 5, in which the polymer which can complex with the starch is selected from copolymers of polyester/polyurethane, polyamide/polyesters, aliphatic and aliphatic aromatic polyesters and polyamides.

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12. Polymers according to claims 1 or 2, wherein the starch complexing agent contains reactive groups for the hydrophobic matrix.



13. Polymers according to any of claim 1 to 12, in which the hydrophobic polymer incompatible with starch is selected from the group consisting of ethylene polymers and copolymers, crystalline propylene polymers and copolymers, aromatic polyester resins, polyamides, polyoxymethylene resins, polyphenylene oxide resins, and polycarbonates.

14. Polymers according to any of claims 1 to 12, in which the hydrophobic polymer is a rubber selected from the group consisting of styrene-butadiene rubbers, polybutadiene rubbers, polyisoprene rubbers, ethylene-propylene and ethylene-propylene-diene rubbers, and natural rubber.

15. A method for preparing filled polymers according to any of claims 1 to 14, in which a composition comprising the starch/polymer complex, forming part of a continuous interpenetrated structure between the complexing polymer and the complex is mixed, in the melt state or under hot mastication conditions, with the hydrophobic polymer incompatible with starch, in the presence of coupling agents containing groups reactive with the polymer matrix and with the complex.

16. A method preparing filled polymers according to any of claims 1 to 14, in which a composition comprising the starch/polymer complex is mixed with a rubber at a processing temperature between 140 and 160°C, in the presence of coupling agents containing groups reactive with the polymer matrix and with the complex.

17. A method according to claims 15 and 16, in which the coupling agent is selected from vinyl and tetrasulphide silanes and alkyl titanates.

18. A method according to any of claims 15, 16 and 17 in which the coupling agent is used in a quantity of from 0.5 to 10% by weight of the complex.



19. A method for preparing filled polymers according to claims 1 to 14, wherein the polymeric matrix is a biodegradable polymer selected from the group consisting of the aliphatic-aromatic polyesters, the aliphatic polyamides, the polyamides-polyesters, polyurethane-polyesters, polyurethane-polyamides and polyurea-polyesters comprising melt-mixing the polymer forming the polymeric matrix with a complex of starch having the characteristics as set forth in claim 1 and further characterised by being formed of starch complexed with a complexing agent different from the polymer forming the matrix and from the ethylene-vinylalcohol copolymers.

20. A method according to claim 19, wherein the starch complex is preformed or formed during melt-mixing.

21. Shaped articles obtainable from the hydrophobic polymers of claims 1 to 14.

22. Shaped articles obtainable from the hydrophobic polymers of claims 1 to 14, wherein the hydrophobic polymer is selected from the group consisting of the aliphatic and aliphatic-aromatic polyesters, polyurethane-polyamides, polyurea-polyesters, and polyurethane-polyesters.

23. Films and compostable bags obtainable from the hydrophobic polymers of claims 1 to 14.

24. Tyres obtainable from the rubbers of claim 14.



PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference PC273PR	FOR FURTHER ACTION		see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.
International application No. PCT/EP 99/ 07038 ✓	International filing date (day/month/year) 22/09/1999 ✓	(Earliest) Priority Date (day/month/year) 22/09/1998 ✓	
Applicant NOVAMONT S.P.A. et al. ✓			

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

1. Basis of the report

a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ **Certain claims were found unsearchable** (See Box I).

3. ☐ **Unity of invention is lacking** (see Box II).

4. With regard to the **title**,

☒ the text is approved as submitted by the applicant. ✓

☐ the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☒ the text is approved as submitted by the applicant. ✓

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the drawings to be published with the abstract is Figure No.

☐ as suggested by the applicant. ✓

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

☐ None of the figures.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 99/07038

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08L29/04 C08L21/00 C08L23/02 C08L67/00 C08L59/00
 C08L71/12 C08L69/00 C08K5/548

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08L C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 92 14782 A (NOVAMONT SPA) 3 September 1992 (1992-09-03) cited in the application page 19, paragraph 6 -page 20, paragraph 2; claims 1,13 ---	1,6,8,9, 20
E	EP 0 965 615 A (NOVAMONT SPA) 22 December 1999 (1999-12-22) claims 1,5,11,12; examples ---	1,12,18
X	WO 98 20073 A (BELLOTTI VITTORIO ;CELLA GIAN DOMENICO (IT); DEL GIUDICE LUCIANO ()) 14 May 1998 (1998-05-14) claims 1,11,28,38,40,48 ---	1,2,4, 12,18,22
A	EP 0 404 727 A (WARNER LAMBERT CO) 27 December 1990 (1990-12-27) claims 1,4,11-13; examples ---	1,4,7,12
-/--		

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

25 January 2000

Date of mailing of the international search report

04/02/2000

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INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 99/07038

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE WPI Derwent Publications Ltd., London, GB; AN 94-057775 XP002031921 & CA 2 069 861 A (BESHAY) 29 November 1993 (1993-11-29) abstract ---	1,12,13
X	EP 0 795 581 A (GOODYEAR TIRE & RUBBER) 17 September 1997 (1997-09-17) page 3, paragraph 3; claims 1,3; examples -----	1,13-16, 23



INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 99/07038

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WO 9214782	A	03-09-1992	IT 1245408 B	20-09-1994
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